

APPLICATION OF ORGANIC TRACERS IN CHARACTERIZING  
THE GREATER CONFINEMENT DISPOSAL TEST AT THE  
NEVADA TEST SITE

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ABSTRACT

The Greater Confinement Disposal Test (GCDT) is a research project investigating the feasibility of augered-shaft disposal of low-level radioactive waste considered unsuitable for shallow land burial. Gaseous diffusion of radionuclides through alluvial sediments is considered the primary contaminant migration process. Volatile halocarbon tracers are released in the subsurface and their migration is monitored to determine media effective diffusion coefficients, tortuosity values, and sorption terms. Design and instrumentation of the emplacement and monitoring shafts of the disposal facility are detailed. Instrumentation includes a three-dimensional array of soil-air sample stations encircling the disposed waste. Recirculation flow lines minimize induced advection in the alluvial matrix due to tracer sample collection.

INTRODUCTION

Low-level radioactive waste with a high-specific activity or long-term potential for migration through the biosphere may require greater confinement than routine shallow land burial facilities afford. Augered shafts, deep trenches, improved waste forms, engineered structures, and hydrofracturing have been proposed as methods for achieving the greater confinement of concentrated, low-level radioactive waste. A generic analysis of these disposal alternatives indicated that augered shafts in the southwestern United States may provide the lowest health risk, at an intermediate cost.<sup>1</sup> In 1981 the U.S. Department of Energy's (DOE) National Low-Level Waste Management Program and the Nevada Operations Office initiated the GCDT augered-shaft demonstration project at the Nevada Test Site. The GCDT project is a full-scale disposal facility containing environmentally mobile and high-specific-activity waste sources.<sup>2</sup>

A research program has been developed to characterize the hydrogeologic environment and to analyze potential contaminant migration processes. Gaseous diffusion through the porous alluvial matrix is expected to be the primary transport process. Volatile organic tracers will be used for *in situ* gaseous diffusion tests to determine media diffusion coefficients, tortuosity values, and sorption terms. The long-term performance of the GCDT will be evaluated through the application of a solute transport numerical model, which will be capable of modeling all radionuclide transport processes of significance at the GCDT. The operational standard for the GCDT will be compliance with the performance objectives detailed in DOE Order 5820.2, "Radioactive Waste Management," for low-level waste, shallow land burial sites. The research program will also include an analysis of several secondary transport processes that may influence gaseous dif-

fusion or offer significant alternative migration pathways.

HYDROGEOLOGIC SETTING

Frenchman Flat, site of the GCDT, is characteristic of the Southwest's Basin and Range physiographic province. Frenchman Flat is a closed basin primarily composed of Quaternary alluvium; at its center is a large playa deposit of silt and clay (Fig. 1). The Massachusetts Mountains to the northwest and Black Ridge to the west are Tertiary volcanic tuffs and ash flows. The mountain perimeter to the east is Paleozoic, mostly undifferentiated carbonate and quartzite formations. Mercury Ridge to the south is a mixture of Tertiary and Paleozoic material. The GCDT rests on a piedmont formed by the coalescence of alluvial fans during the Pleistocene. The alluvial material was derived from the Tertiary volcanics of the Massachusetts Mountains.

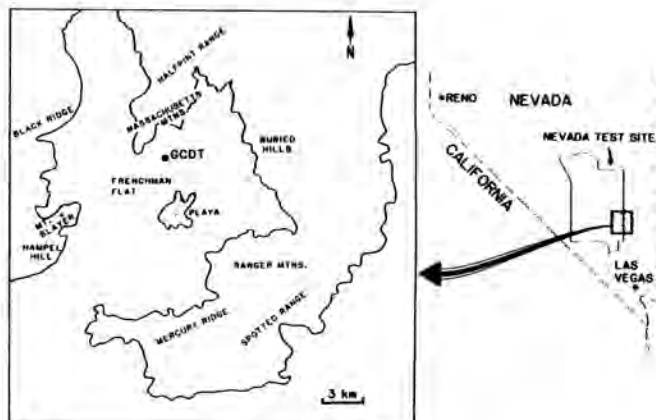


Fig. 1. General location map of the GCDT.

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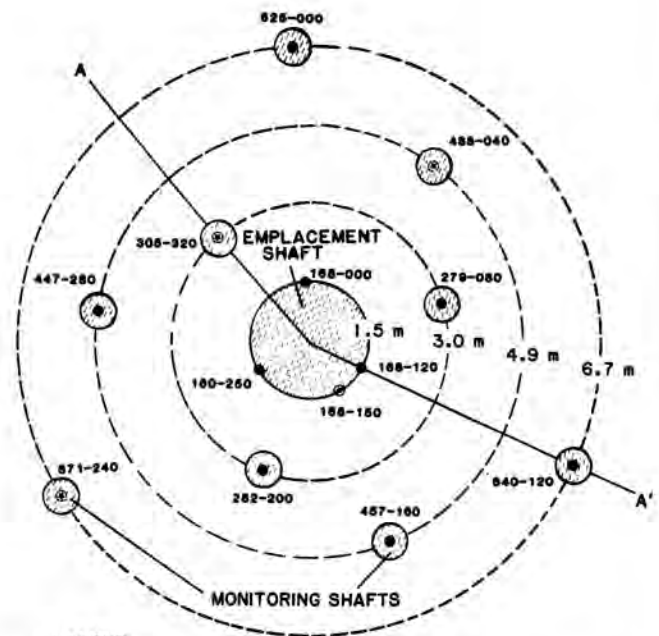
The groundwater system below the GCDT is part of the Ash Meadows aquifer. This regional aquifer is an interbasin flow system in the basement Paleozoic carbonates which is relatively independent of the topographic boundaries of Frenchman Flat. Below the GCDT, the Ash Meadows aquifer occupies the lower portion of the Cenozoic alluvial fill. The direction of groundwater flow under the GCDT is to the south southwest. Depth to water at the study site is approximately 245 m.

The soil at the GCDT developed in a high temperature, low rainfall environment. The soil is a coarse-textured sandy loam with a low organic content. The soil moisture varies between 4 and 8 percent of the matrix pore volume. Inspection of the alluvial material during drilling of the emplacement and associated monitoring shafts revealed the soil to be relatively homogeneous. A few lenses of coarser material were the only notable exceptions.

#### FACILITY DESIGN

The GCDT consists of a central emplacement shaft and nine monitoring shafts below a 20-m by 20-m concrete pad. The emplacement shaft is 36 m deep and 3 m in diameter. The monitoring shafts, which are the same depth but only 60 cm in diameter, are orbitally staggered at radii of 3, 4.9, and 6.7 m from the central shaft. (See Fig. 2.)

The GCDT has nine instrument lines with monitoring stations, each of which have a soil atmosphere sampler, a thermocouple psychrometer, and an independent thermocouple (Fig. 3). The emplacement shaft has a 5.5-m-tall tripod centered at the bottom, which was used to position three instrument lines along the perimeter of the emplacement shaft. The other six lines are in monitoring shafts. On



#### LEGEND

- ⊙ ACCESS TUBE FOR NEUTRON SCATTER PROBE
- INSTRUMENT STRING WITH SOIL AIR SAMPLERS, THERMOCOUPLES, & THERMOCOUPLE PSYCHROMETERS
- ▣ SIFTED ALLUVIAL BACKFILL

NOTE LOCATION DESCRIPTOR: POINT 279-080 IS AT A RADIUS OF 290 cm AND AN AZIMUTH OF 80°

Fig. 2. Positioning of the GCDT emplacement and monitoring shafts.

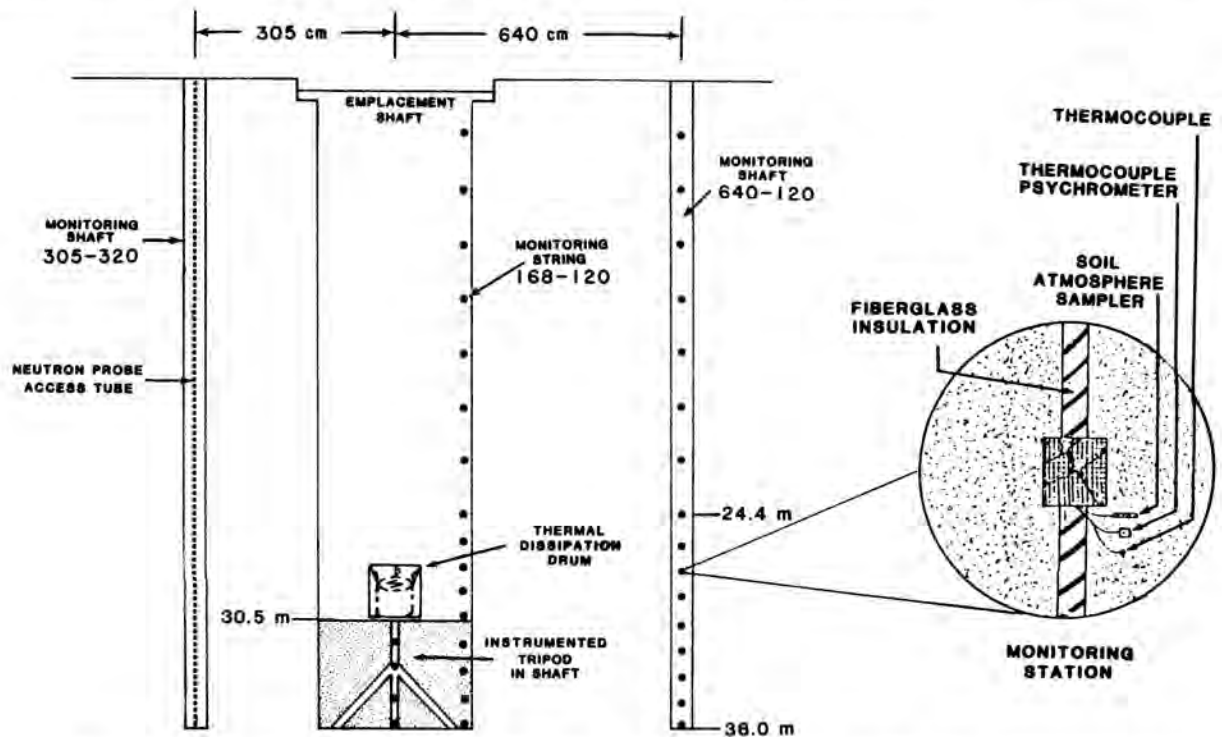


Fig. 3. GCDT cross-sectional view through transect A-A' in Fig. 2 showing instrumentation.

each line, monitoring stations are spaced every 3 m down to a depth of 24 m, and every 1.5 m thereafter; totaling sixteen stations per instrument line. Four monitoring stations are included on the tripod. Three 5-cm-diameter, 36-m-long aluminum access tubes are reserved as access ports for a neutron scatter probe.

After the instrument lines and the neutron and gamma access tubes were emplaced, the monitoring shafts were completely backfilled with sifted alluvium. The emplacement shaft was backfilled to the 30.5-m depth to cover the tripod before any waste was added. The remainder of this shaft was backfill as the waste was emplaced.

The GCDT project also includes a 6-m-deep shallow test plot (STP). The STP design is similar to the GCDT, has the same types of instrumentation, and shares the same analytical and computer facilities. The STP has served as a calibration center for developing reliable operational procedures for analytical, mechanical, and computer equipment at the GCDT. A rheostatically controlled, 2000-W electrical heat source is centered in the STP to permit simulation of GCDT temperatures. Thermocouples inside and on the outer surface of the heat source monitor thermal output. The STP was completely stemmed with sifted alluvial fines after installation of the monitoring instrumentation and heat source. Experience gained at the STP in the integrated system's performance is critical to the reliability of the organic tracer diffusion tests at the GCDT.

#### EMPLACED WASTE

Between December 15, 1983, and March 12, 1984, high-specific-activity and environmentally mobile sources were emplaced in the GCDT shaft in several stages or levels. These levels are differentiated by source material and backfill interval (Fig. 4). To avoid macroscopic void spaces during backfilling, sifted alluvium was poured downhole at appropriate intervals as dictated by the form or quantity of deposited waste.

Eight unshielded, high-specific-activity Sr-90, Cs-137, and Co-60 sources were placed in a thermal dissipation drum located in level B. Other high-specific-activity Sr-90, Ra-226, and Ac-227 sources were placed in level C. Levels D through F of the GCDT contain tritium in seventy-six 210-L drums. Levels A and G only contain sifted alluvium. Total activity of the disposed waste in the GCDT shaft was 1.11 MCI as of May 1, 1984. Table I shows the distribution of the waste in the GCDT emplacement shaft.

#### INSTRUMENTATION

Determination of the gaseous diffusion characteristics of the GCDT alluvial matrix is dependent on the ability to monitor the diffusive flux of minute quantities of volatile halocarbon tracers. The basic experimental procedure involves releasing a known quantity of a gaseous compound at a known location in the porous medium and measuring its diffusive migration past multiple observation points. The three-dimensional array of 144 GCDT monitoring stations can each be used to retrieve soil-air samples, measure ambient temperatures and soil moisture potential.

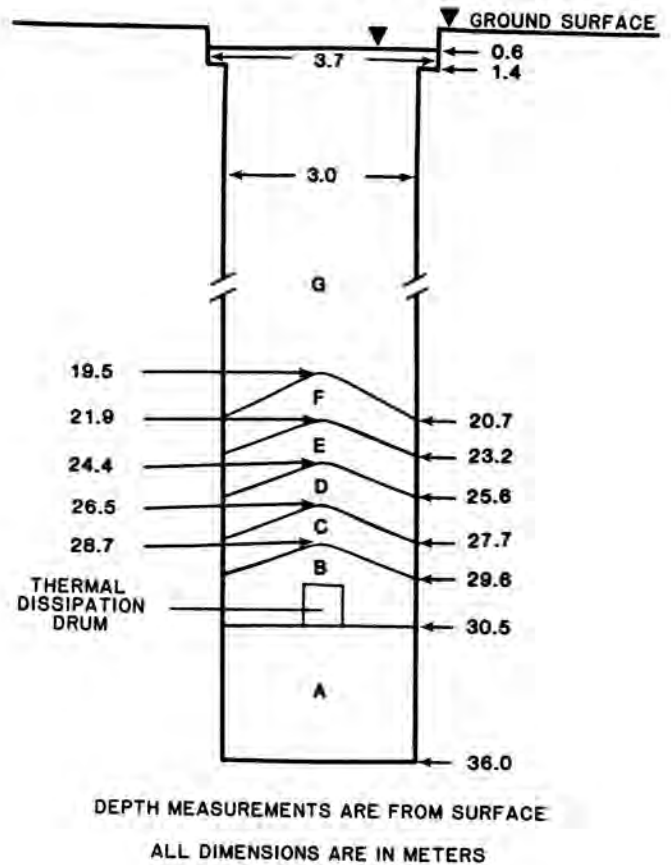


Fig. 4. Stemming levels in the GCDT emplacement shaft.

TABLE I  
Waste Distribution in GCDT Emplacement Shaft

Depth Level <sup>a</sup>	Radio-nuclides	Activity <sup>b</sup> (Ci)	Thermal Output <sup>b</sup> (W)
A	none	----	----
B	Sr-90, Cs-137 and Co-60	476,960	3,165
C	Sr-90, Ra-226 and Ac-227	40,170	273
D	H-3	152,090	5
E	H-3	266,160	9
F	H-3	175,280	6
G	none	----	----

<sup>a</sup>See Fig. 4.

<sup>b</sup>Activity and thermal output as of May 1, 1984.

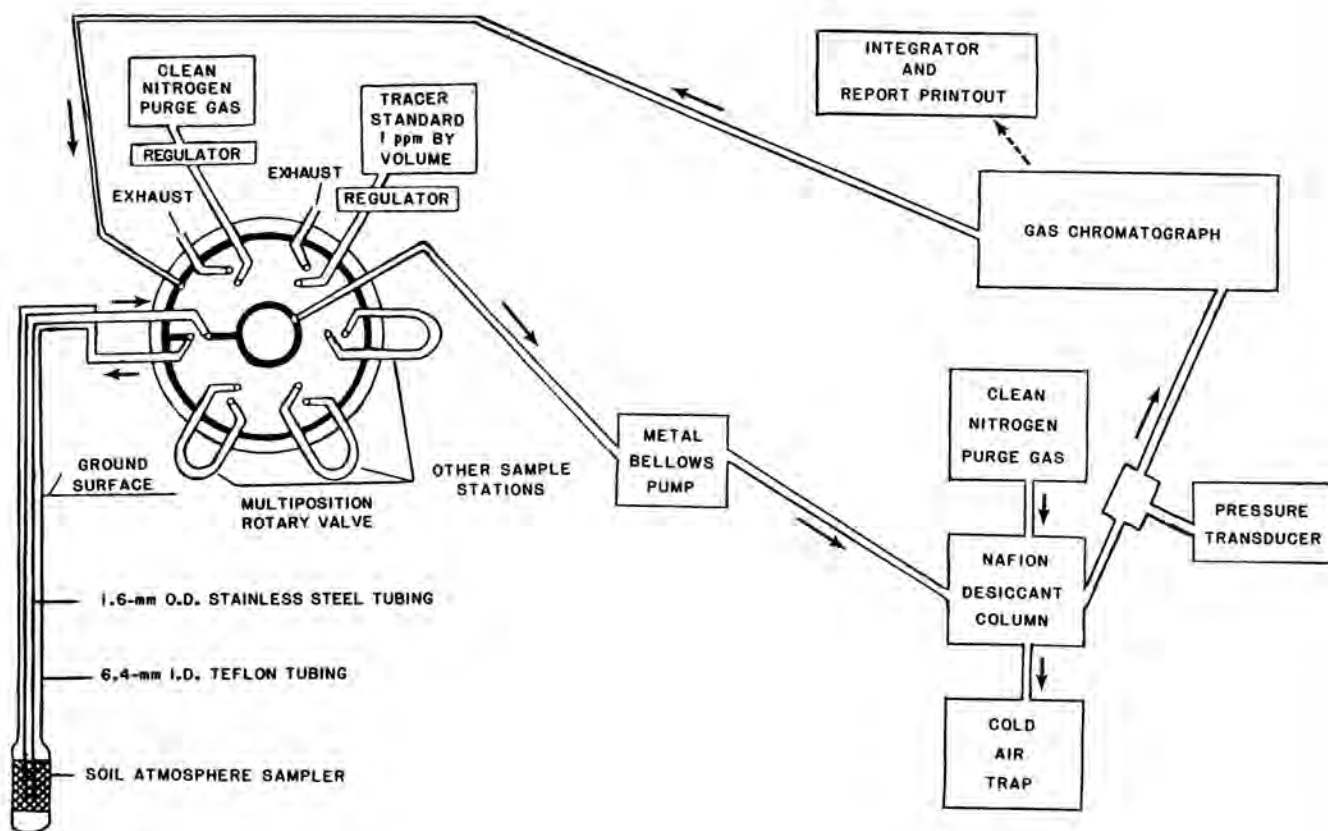


Fig. 5. Schematic of GCDT system for analyzing soil-air samples.

Soil-air samples are retrieved via a metal bellows pump drawing through a stainless steel sample line. Sorption of organic tracers on stainless steel is insignificant. Soil air is recirculated downhole through Teflon tubing, the annulus of which contains the stainless steel sample line. Soil air is recirculated to minimize or eliminate convective flow due to sample collection. The Teflon and stainless steel tubing terminate downhole in a soil-air sampler. At the surface, the recirculation sample lines feed into electronically actuated, air-driven multi-position rotary valves. The multi-position valves facilitate selection of and switching between monitoring stations. Tracer concentrations are analyzed using a gas chromatograph equipped with electron-capture detectors capable of identifying halogenated compounds in parts per billion. (See Fig. 5.)

Soil-air from an activated sample line flows from a multi-position valve to the gas chromatograph, passing through a desiccant column and past a pressure transducer which is positioned between the pump and the gas chromatograph. During pumping of soil-air samples through the recirculation sample line, soil-air merely passes through the injection valve sample loop of the gas chromatograph and returns downhole. After an appropriate pumping interval, unique to each sample station, the pump is deactivated and pressure in the sample line is monitored by the in-line transducer. A soil-air sample is injected to the gas chromatograph when pressure in the sample line equilibrates with atmospheric pressure. The soil-air sample stations can also serve as tracer release point sources or air piezometers.

Organic tracers contained in permeation tubes are placed inside a modified condensation tube connected to a subambient circulating water bath. Permeation tubes are typically small Teflon containers that are initially 70 to 90 percent filled with volatile halogenated organic liquid. Tracer is released by diffusion through the Teflon walls of the container. Tracers with high vapor pressures are contained in steel ampules with Teflon excursion ports. Diffusion through the Teflon of these devices is temperature dependent, and can also be adjusted by the choice of Teflon surface area and thickness. The tracer release rate is controlled by adjusting the water bath temperature. A metal bellows pump draws soil air through the outer Teflon tubing, past the condensation tube containing tracer permeation devices, and returns it downhole through the stainless steel inner tubing to the soil-air sampler, where the tracer diffuses through the alluvial matrix. The soil-air sampler thus serves as a temperature-controlled point source.

#### ORGANIC TRACER DIFFUSION

The gaseous diffusion of volatile halocarbon tracers through the porous matrix at the GCDT is being measured to quantify media diffusion coefficients, tortuosity values, and tracer sorption terms under both isothermal and thermally graded conditions. Volatile halocarbons are useful tracers because they can be detected in minute concentrations and do not alter the diffusive characteristics of the medium. In addition, volatile halocarbons are uncommon in the environment (with notable exceptions), chemically stable, and inexpensive.

TABLE II

Halocarbon Tracers for GCOT Experiments

Compound	Name	Molecular Weight (g)	Boiling Temp. (°C)	Critical Temp. (°C)
CHClF <sub>3</sub>	F22	80.47	-40.75	96.0
CBrF <sub>3</sub>	F13B1	148.92	-57.75	67.0
CClB <sub>r</sub> F <sub>2</sub>	F12B1	165.36	-3.98	153.9
CCl <sub>2</sub> F <sub>2</sub>	F12	120.91	-29.79	110.0
CCl <sub>3</sub> F	F11	137.37	+23.82	198.0

The nontoxic halocarbons currently being utilized as tracers in the GCOT experiments are listed in Table II.

Analysis of gaseous diffusion of tracers through porous media is based on Fick's Second Law, a mass balance equation describing one-dimensional, nonsteady-state diffusion of one gas into another:<sup>3</sup>

$$\frac{\partial c_A}{\partial t} = D_{AB} \frac{\partial^2 c_A}{\partial x^2} \quad (1)$$

where  $D_{AB}$  = molecular diffusion constant for diffusion of gas A into gas B (cm<sup>2</sup>/s),  
 $c_A$  = concentration of gas A (mol/cm<sup>3</sup>),  
 $x$  = dimension in the direction of diffusion (cm), and  
 $t$  = time (seconds).

The given relation assumes a constant density, isothermal environment and does not account for convection of the gas or reactive interchanges. An implied assumption is that  $D$  is constant for a given medium, temperature, and pressure.

In partially saturated porous media, Fick's Second Law may be generalized to account for the reduction in cross-sectional flow area, potential sorption phenomena, and the matrix tortuosity. Assuming that an immobile liquid phase completely wets the matrix, and that rapid equilibration occurs between the gas phase and the dissolved and sorbed gas in the liquid and solid phases, an effective diffusion coefficient ( $D'$ ) can be defined:

$$D' = \frac{\tau \eta_g D_{AB}}{\eta_g + (\eta_t - \eta_g) \rho_w k_w + (1 - \eta_t) \rho_s k_s} \quad (2)$$

where  $\tau$  = the tortuosity factor (dimensionless),  
 $\eta_g$  = drained or gas-filled porosity (dimensionless),  
 $\eta_t$  = total porosity (dimensionless),  
 $\rho_w$  = density of water (g/cm<sup>3</sup>),  
 $\rho_s$  = particle density of granular material making up solid matrix (g/cm<sup>3</sup>),  
 $k_w$  = liquid-gas partitioning coefficient (moles/g water ÷ moles/cm<sup>3</sup> gas),  
 $k_s$  =  $k_p k_d$  = gas-liquid-solid distribution product (cm<sup>3</sup> gas/g solid), and

$k_d$  = solid-liquid distribution coefficient (moles/g solid ÷ moles/g water).

The effective diffusion coefficient is unique for different media and gases because it is a "lumped" parameter of subcomponent variables such as structural and sorptive properties of the system. The denominator of Eq. 2 may be considered a sorption term, the media sorption-corrected porosity ( $A$ ):<sup>4</sup>

$$A = \eta_g + (\eta_t - \eta_g) \rho_w k_w + (1 - \eta_t) \rho_s k_s \quad (3)$$

Thus, the partial differential governing equation describing gaseous diffusion in an isotropic, homogeneous, and partial saturated porous medium under isothermal conditions is:

$$\frac{\partial c_A}{\partial t} = D' \frac{\partial^2 c_A}{\partial x^2} \quad (4)$$

where

$$D' = \frac{\tau \eta_g D_{AB}}{A} \quad (5)$$

Fick's Second Law of gaseous diffusion is analogous to equations describing heat conduction in solids (Fourier's Law) and flow of fluids in porous media (Darcy's Law). Solutions for these equations are abundant in the literature for various source configurations and boundary conditions.

Carslaw and Jaeger describe a three-dimensional solution for temperature at any point and time, given a continuous point source of heat in an infinite medium.<sup>5</sup> Substituting equivalent gaseous diffusion parameters for heat flow terms, the solution of Eq. 4 in radial coordinates is:

$$c = \frac{q}{4\pi D' r A} \operatorname{erfc}(r/\sqrt{4D't}) \quad (6)$$

where  $\operatorname{erfc} = 1 - 2/\sqrt{\pi} \int_0^x \exp(-z^2) dz$

$q$  = constant tracer mass production rate,  
 $t$  = time period of mass production, and

$$r = \sqrt{(x-x_0)^2 + (y-y_0)^2 + (z-z_0)^2}.$$

As time reaches infinity, Eq. 6 reduces to  $c = q/4D'rA$ , representing a steady-state distribution of tracer concentration in which a constant supply of tracer gas is introduced at a central point and diffuses outward into the infinite medium. The continuous point source solution (Eq. 6) is the basis of tracer diffusion tests at the GCOT.

Equation 6 can be solved by least squares analysis or a graphical super-position technique.<sup>3</sup> Field data consists of tracer concentration versus time curves at a given radial distance from the point source. Once the effective diffusion coefficient ( $D'$ ) and sorption term ( $A$ ) are quantified, tortuosity can be solved for in Eq. 5 if the general

diffusion coefficient is known. Boundary conditions applied to Eq. 6 refine the model. At the GCDT, the ground surface is considered a zero concentration boundary and is mathematically simulated as a tracer sink equidistant from the source. The water table may be modeled as a zero flux boundary, or neglected due to the great depth to water at the GCDT.

Greater vapor flux may occur at the GCDT due to a thermal gradient generated by the buried waste. Fick's Second Law may be modified with the inclusion of mechanistic or phenomenological "enhancement factors".<sup>6</sup> The GCDT project will include a numerical model that will include an analytical expression to compute a thermal enhancement factor.

#### STATUS OF THE GCDT PROJECT

In situ tracer diffusion tests at the GCDT are scheduled for mid-1985, pending completion of the GCDT facilities. Operational tests of the soil-air sampling system have been conducted at the STP calibration facility. These tests have yielded valuable information on minimizing line turbulence, absorption, and pressure and temperature differentials in the recirculation sample lines. Sampling procedures and calibration curves have already been developed which will greatly increase the reliability of the diffusion tests at the GCDT. Isothermal and simulated thermally graded diffusion tests will be done first at the STP, followed by large scale in situ tests in the GCDT in late 1985 and continuing through 1986.

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